TEGRAL MEASUREMENTS IN EBR-II OF PTURE RATES, FISSION RATES, AND ALPHA R ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, AND ^{242}Pu

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INTEGRAL MEASUREMENTS IN EBR-II OF CAPTURE RATES, FISSION RATES, AND ALPHA FOR ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, AND ^{242}Pu

by

R. R. Heinrich, J. Williams, A. A. Madson,
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Chemical Engineering Division

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ABSTRACT

This report describes a study, performed over a five-year period, in which integral capture rates, fission rates, and their ratio, alpha, have been determined for ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, and ^{242}Pu as a function of position in EBR-II. A total of 64 samples were irradiated in EBR-II over a period of 6 to 12 months for accumulated exposures ranging from 2000 to 10,000 MWd. Fission rates were determined by assaying the fission product ^{137}Cs, and capture rates were determined by mass-spectrometric and alpha-spectrometric analysis. Breeding potentials for the six isotopes have been calculated from the alpha values determined in the core and blanket of EBR-II.

I. INTRODUCTION

One of the most critical and elusive quantities necessary to the design and development of large breeder reactors is the capture-to-fission ratio, alpha. This quantity influences both the economics (breeding potential) and the design and safety (Doppler coefficient) aspects of breeder reactors. Measurement of capture-to-fission ratios for heavy elements has been the objective of a number of recent experiments;[1-6] primary emphasis has been upon differential measurements of ^{239}Pu. Although differential measurements are essential to reactor design, integral foil-activation measurements offer a unique means for testing design parameters by direct measurement within an operating power reactor.

Even before the publication of the recent experiments, it was obvious that discrepancies existed in the available data for heavy-element capture and fission rates. Accordingly, in 1964, we undertook a program to measure integral capture and fission rates throughout the core and blanket of the Experimental Breeder Reactor-II (EBR-II). This report describes the study, performed over a seven-year period, in which integral capture rates, fission rates, and alpha values have been determined for ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, and ^{242}Pu as a function position in EBR-II. A total of 64 samples were irradiated in EBR-II over a period of 6 to 12 months for accumulated exposures ranging from 2000 to 10,000 MWd. The number of fissions occurring in the samples was determined by assaying the fission product ^{137}Cs, and the capture products were measured by mass-spectrometric and alpha-spectrometric analysis.

The intent of this report is to provide a complete description of the experimental aspects of these measurements and to summarize the large amount of data obtained. Additional reports, in which these results are compared to reactor-physics calculations, will be published at a later date.

II. EXPERIMENTAL

A. Irradiations of Samples

The Experimental Breeder Reactor-II (EBR-II) operates with fast neutrons and is cooled with liquid sodium. During our irradiations, the central core generally consisted of fuel rods containing 49% enriched ^{235}U assembled in 67 subassemblies. The inner and outer blankets consisted of rods of depleted uranium which were also cooled with liquid sodium. The reactor was designed to operate at 62.5 MW(t) with a full-power neutron flux at core center of 6.9×10^{14} n/(cm^2)(sec) having energies greater than 1.35 MeV and 2.9×10^{15} n/(cm^2)(sec) having energies less than 1.35 MeV. The irradiations reported here were performed in reactor power Runs 5 thru 23, in which the maximum operating power was 45 MW(t). Further details on the construction of EBR-II and its fuel and blanket elements can be obtained in Refs. 7 and 8.

A total of 64 uranium and plutonium samples were irradiated in 8 subassemblies; 3 in the core, 1 in the inner blanket, and 4 in the outer blanket. Each isotopic sample was contained in a Type 304 stainless steel capsule which varied in size depending upon the type of fuel element into

which the sample was placed. Each capsule was capped, welded, and then helium leak-tested before being loaded into its respective element. Elemental composition of the Type 304 stainless steel capsules is given in Table 1. Core capsules were tubes 0.75 in. long with a diameter of 0.14 in. and a wall thickness of 0.015 in. and were bonded to the elements with sodium. The upper blanket capsules, located in core subassemblies, were tubes 0.625 in. long by 0.234-in. diameter. These capsules were contained in a secondary container which was also bonded to the element with sodium. The inner and outer blanket capsules were 0.537 in. long by 0.359 in. diameter and like the upper blanket capsules were secondarily contained and bonded with sodium. Wall thicknesses of the blanket capsules were 0.015 in. Capsule weights ranged from 1-4 g. The distribution of samples within the reactor and the exact positions of these samples in terms of distance from the central axis and the mid-plane of the reactor are given in Table 2. A typical layout is illustrated by the ^{239}Pu distribution in Fig. 1. Figure 2 represents the horizontal section of the reactor and illustrates the subassemblies which contained the samples. The irradiation was carried out over 24 power runs, and this sector of the reactor was reserved solely for our samples to minimize any perturbations of the neutron flux.

The samples of uranium isotopes were loaded into their respective capsules as a stoichiometric oxide, U_3O_8. The ^{238}U material was originally received as a metal; it was dissolved in HNO_3, taken to dryness, and then converted to U_3O_8 by heating in air at 950°C for several hours. The ^{235}U and ^{233}U isotopes were received as U_3O_8 and no preparation was required before loading into the capsules. All three uranium isotopes were weighed as U_3O_8 by difference in their respective capsules using a microanalytical balance. The amount of U_3O_8 in each capsule ranged from 1 to 3 mg.

The plutonium isotopes, unlike the uranium isotopes, were loaded into the capsules as a wet nitrate. An aliquot of the plutonium nitrate solution of unknown concentration was pipetted into the respective capsules and allowed to dry in a desicator before the capsules were capped and welded. Final concentration in the capsule was approximately 0.5 mg. The exact plutonium concentration of each isotope was determined after the irradiation by methods described below.

At the time when the uranium and plutonium samples were being encapsulated, control samples were also prepared. These controls were of approximately the same concentration of uranium or plutonium as the irradiation samples and were capped and welded identically. The controls were then stored until after the irradiation and were analyzed identically to their appropriate counterparts. Isotopic compositions of the samples before irradiation are given in Table 3.

B. Analysis of Samples

The analytical procedure for each isotope is discussed in detail below. However, a number of standard procedures were used on all samples and these are discussed in general here.

After irradiation, the capsules were removed from the subassemblies and allowed to cool for approximately one year. Before dissolution, the capsules were washed with ethyl alcohol and water to remove any excess

TABLE 1. Elemental Composition of Type 304 Stainless Steel Capsules

Element	Concentration (wt %)
Iron	70.6
Chromium	18.5
Nickel	9.43
Manganese	0.96
Silicon	0.28
Cobalt	0.10
Copper	0.08
Nitrogen	0.043
Phosphorus	0.016
Sulfur	0.015

TABLE 2. Summary of Irradiation-Location Data for Alpha Measurements

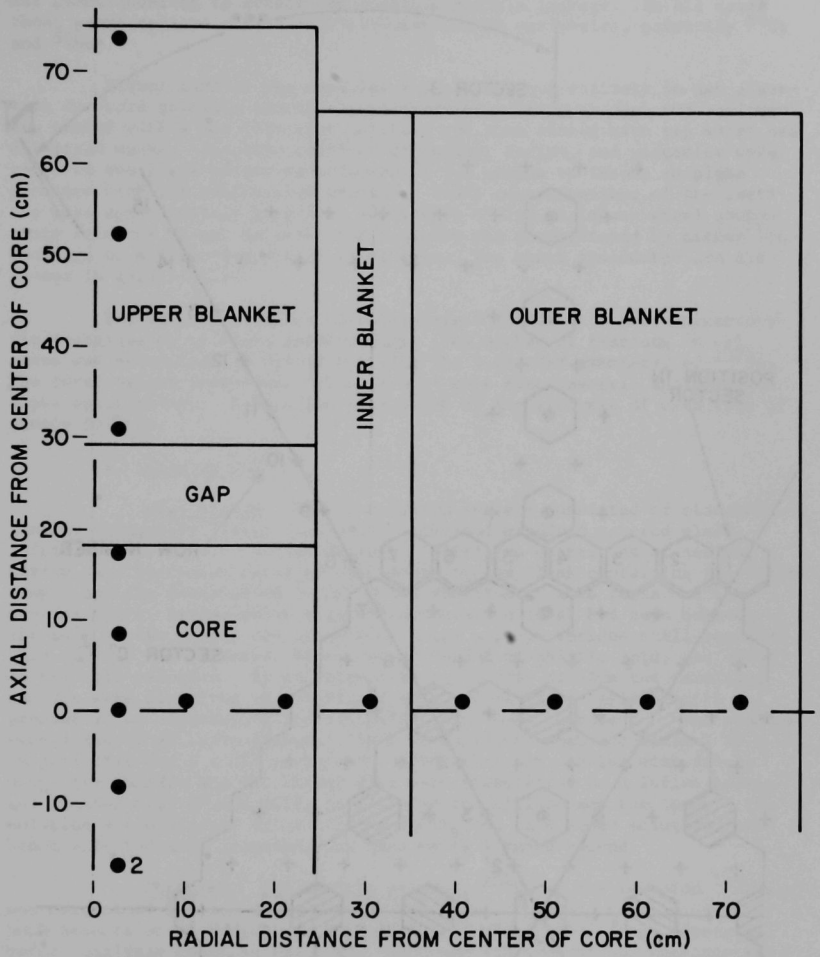
Grid Position	Subassembly Number	Radial Position (cm)	Axial Position (cm)	Sample Number ^{233}U	^{235}U	^{238}U	^{239}Pu	^{240}Pu	^{242}Pu	Date In	Date Out
1-A-1	C220X	2.86±0.14	-17.0±0.5	1	16	26	36	–	–	7/29/65	1/22/66
		"	-8.5±0.5	2	14,15	–	35	–	61		
		"	+8.5±0.5	4	–	25	31	51	59		
		"	+17.0±0.5	5	12	24	30	–	58		
		"	0.0±0.5	7	13	–	32	48	60		
		2.50±0.26	+72.9±0.5	–	20	–	41	54	67		
		"	+51.5±0.5	8	–	–	73	53	66		
		"	+30.2±0.5	–	19	–	39	52	65		
3-B-2	C221X	10.21±0.14	+2.22±0.5	–	–	–	–	49	–	7/29/65	1/22/66
		"	+0.32±0.5	–	–	–	33	–	–		
		"	-1.59±0.5	–	–	–	–	–	64		
		"	-3.49±0.5	–	17	–	–	–	–		
5-B-2	C222X	21.25±0.14	+3.2±0.5	6	–	–	–	–	–	10/13/65	5/23/66
5-B-4		"	+2.22±0.5	–	–	–	–	50	–		
		"	+0.32±0.5	–	–	–	34	–	–		
		"	-1.59±0.5	–	–	–	–	–	63		
7-B-4	A776X	30.62±0.4	-0.94±0.5	–	–	–	–	–	68	5/12/65	10/21/66
		"	+0.42±0.5	–	–	–	42	–	–		
		"	+1.78±0.5	–	–	–	–	55	–		
		"	-2.30±0.5	–	21	–	–	–	–		
9-B-5	U1548X	40.83±0.4	+1.78±0.5	9	–	–	–	56	–	5/12/65	8/7/66
		"	+2.30±0.5	–	–	–	–	–	–		
		"	-0.94±0.5	–	–	–	–	–	69		
		"	+0.42±0.5	–	–	–	43	–	–		
11-B-6	U1549X	51.03±0.4	-0.94±0.5	–	–	–	–	–	70	5/12/65	8/7/66
		"	+0.42±0.5	–	–	–	44	–	–		
		"	+1.78±0.4	–	–	28	–	–	–		
		"	-2.30±0.5	–	22	–	–	–	–		
13-B-7	U1550X	61.24±0.4	+1.78±0.5		–	–	–	57	–	5/12/65	12/3/66
		"	-2.30±0.5	10	–	–	–	–	–		
		"	-0.94±0.5	–	–	–	–	–	71		
		"	+0.42±0.5	–	–	–	45	–	–		
15-B-8	U1551X	71.45±0.4	-0.94±0.5	–	–	–	–	–	72	5/12/65	12/3/66
		"	+0.42±0.5	–	–	–	46	–	–		
		"	+1.78±0.5	–	–	29	–	–	–		
		"	-2.30±0.5	–	23	–	–	–	–		
Total Number of Irradiated Samples				9	11	5	15	10	14		

TABLE 3. Sample Compositions Before Irradiation

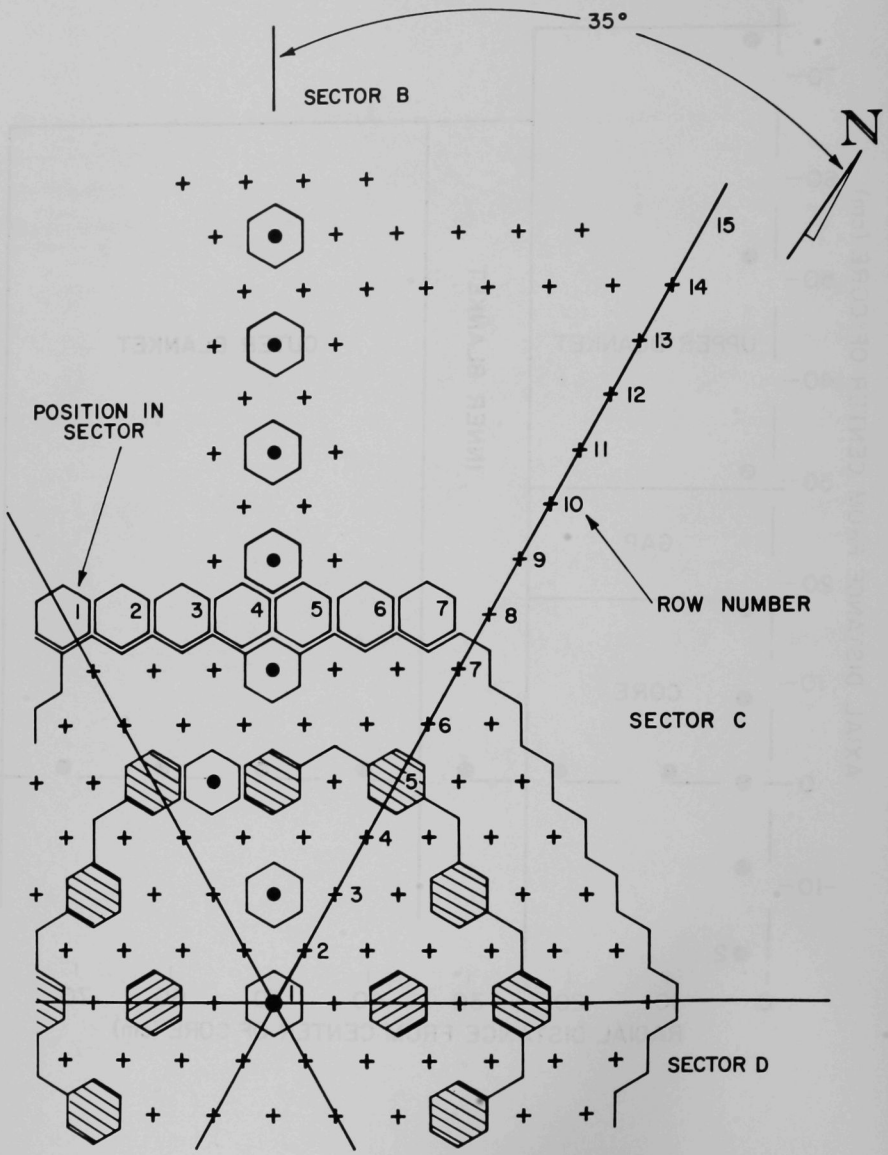
Uranium						
	Composition (at. %)					
Major Isotope	232	233	234	235	236	238
233	$<1\times10^{-4}$	>99.996	$<2\times10^{-4}$	-		$<1\times10^{-4}$
235	-	-	$<1\times10^{-3}$	99.996	2.4×10^{-3}	$<9\times10^{-4}$
238	$<3\times10^{-4}$	$<3\times10^{-4}$	$<3\times10^{-4}$	$<3\times10^{-4}$	$<3\times10^{-4}$	>99.999

Plutonium					
	Composition (at. %)				
Major Isotope	238	239	240	241	242
239	-	>99.996	3.65×10^{-3}	$<2\times10^{-4}$	-
240	-	3.56×10^{-3}	99.614	3×10^{-4}	-
242	0.726	0.230	7.23	3.29	88.53

Figure 1. Distribution of ^{239}Pu Samples in EBR-II



ure 2. Horizontal Section of EBR-II



sodium, and then washed in dilute nitric acid with ultrasonic agitation. These HNO_3 wash solutions were monitored for fission-product activity by NaI gamma-counting to detect any possible capsule leakage. In all cases these gamma spectra showed only stainless steel activities, primarily ^{60}Co and ^{54}Mn.

Dissolution of the capsules was carried out entirely in new glassware to avoid possible isotopic contamination. Prior to use, the equipment was washed with a hot detergent solution and then rinsed with tap water and deionized water. Inactive carriers of cesium, cerium, and zirconium were added to the flask before dissolution of the sample to insure complete exchange with the radioactive product. Final concentrations of the carriers were approximately 1 μg/ml. Separation of the stainless steel components from the target material and products was accomplished by either ion-exchange or solvent-extraction techniques. The exact procedures are discussed in detail below.

The capture products were determined either by a mass spectrometric technique or by alpha spectrometry. The number of fissions in all cases was determined by Ge(Li) counting the 0.662 MeV gamma-ray of ^{137}Cs. The total target atoms were determined by mass spectrometric analysis and alpha spectrometry. A detailed discussion of the analysis of each type of sample follows.

1. Uranium-233

Dissolution. The dissolution vessels consisted of standard tapered Erlenmeyer flasks fitted with extended standard tapered glass joints which served as air condensers. Microgram quantities of cesium, cerium, and zirconium carriers were added to the flask containing the first sample and the dissolution begun by the addition of aqua regia. After several hours, during which time the dissolution flask had been heated, the bulk of the sample was dissolved, but a silicon residue still remained. This residue was filtered, dissolved in fuming perchloric acid, and added to the main solution. In an attempt to avoid this problem the remaining samples were dissolved in perchloric acid. However, a residue still was present after dissolution and filtration again was required. These samples were filtered on Lexan (General Electric Nuclepore Membrane Filter) of 8 μm pore size and 1×10^5 pores/cm^2. After thorough washing with dilute HNO_3, the residue and the filter disk were transferred to a Teflon beaker and treated with HF and $HClO_4$ to dissolve the silicon and the Lexan. The solution was heated to drive off the H_2SiF_6 and this clear solution, combined with the main solution, was made up to a known volume.

Captures. The capture product, ^{234}U, of the reaction $^{233}U(n,\gamma)$ was determined by mass spectrometric analysis. However, because of the bulk amounts of stainless steel present, the uranium had to be separated before analysis could be realized. This was achieved by ion-exchange separation using the method developed by Korkish and Hazan.[9] A portion of the uranium-stainless steel solution was taken to dryness to remove the perchloric acid, and the uranium was adsorbed onto Dowex-1 anion resin from a 9:1 mixture of 2-methoxyethanol and 6N HCl. With this procedure, the bulk of the stainless steel components pass through the column but cobalt,

manganese, and a small amount of iron are also adsorbed. Cobalt and manganese were removed by washing with an 8:2 mixture of the original reagents. The uranium was finally removed by eluting the column with 1N HCl. This technique produced excellent separation of the uranium, but required a large elutriation to remove residual iron, which resulted in a uranium yield of only 50%. The total number of ^{234}U atoms present at the end of the irradiation was determined from the atom ratios measured mass spectrometrically and from the final number of uranium atoms (in this case, the initial number of uranium atoms less the number of atoms fissioned). The number of capture atoms was determined by difference--the number of ^{234}U atoms at the end of the irradiation less those present initially.

Fissions. The number of fissions were determined by counting the fission product ^{137}Cs with a high resolution Ge(Li) gamma-ray spectrometer. An aliquot of the uranium solution was mounted directly for counting of the 0.662 MeV gamma ray with a 10 cm^3 Ge(Li) detector coupled to a 4096 channel analyzer. The detector resolution was 1.6 keV (FWHM) for the 0.662 MeV gamma ray of ^{137}Cs; the detector was calibrated for energy and relative efficiency using the gamma rays of ^{152}Eu. These efficiencies were made absolute by normalizing to absolutely calibrated standards of ^{57}Co, ^{137}Cs, ^{54}Mn, and ^{60}Co. The 0.662 MeV peak activity was converted to disintegrations using a branching ratio of 0.85. In-pile decay corrections and out-pile decay corrections to the end of the irradiation were made for the ^{137}Cs activity using a 30.0 year half-life. Fissions were calculated using the measured ^{137}Cs atoms and the yield of cesium from ^{233}U fission.[10] Appropriate fission yields of ^{137}Cs[10] for all fissioning isotopes are listed in Table 4.

Target Atoms. The number of target atoms initially present were calculated from the accurately weighed amounts of uranium loaded into the capsules before irradiation. The target atoms present at the end of the irradiation are represented by the initial atoms less the atoms captured and fissioned. Results summarized in Tables 5-10 were calculated on the basis of atoms of target material at the beginning of the irradiation.

2. Plutonium-239

Dissolution. Although perchloric acid had been used successfully in the dissolution of the ^{233}U samples, the large quantities of $HClO_4$ in the solutions made the subsequent chemical separations tedious. Also, at times the dissolution was difficult to control and large glassware was required to prevent losses due to spattering. This, in turn required larger volumes for washing and transferring to the volumetric flask. For these reasons, a mixture of aqua regia and perchloric acid was used in dissolving the ^{239}Pu samples. Again, microgram quantities of cesium, cerium, and zirconium carriers were added before the start of dissolution. As before, a silicon precipitate remained after dissolution and was treated in the same manner as described previously. After dissolution of the silicon and the Lexan filter disk, a black residue still remained which resisted treatment with HCl, HNO_3, HF, and $HClO_4$. The residues from all samples were filtered and analyzed by spark-source mass spectrometry. The main constituent was identified as carbon. The residues were also mounted

TABLE 4. Fission Yields of ^{137}Cs[10]

Isotope	^{137}Cs Fission Yield (%)
^{233}U	6.75
^{235}U	6.20
^{238}U	6.30
^{239}Pu	6.58
^{240}Pu	6.30
^{242}Pu	6.30

subsequent gamma counting. Insignificant amounts of Cs activity were present and the residues were then discarded.

Captures. The separation of the plutonium from its stainless steel constituents was accomplished by solvent extraction, and the ^{240}Pu capture product analyzed mass spectrometrically. Aliquots of the plutonium solution were taken to dryness and the residue dissolved in nitric acid. The plutonium was extracted into methyl isobutyl ketone from an acid-deficient $Al(NO_3)_3$ medium. The organic phase was separated, evaporated to dryness, and submitted for mass spectrometric analysis. From the measured atom ratio and the measured final atoms of ^{239}Pu, the total number of ^{240}Pu atoms present at the end of the irradiation were determined. Total capture atoms were determined by difference--total final ^{240}Pu atoms less the ^{240}Pu atoms present initially.

Fissions. When the time came for determining the number of fissions, the high-resolution Ge(Li) detector system used in counting the ^{137}Cs from the ^{233}U samples was unavailable. Chemical separation of the cesium from other fission products was required, and the method chosen was the precipitation of cesium as a perchlorate. A known amount of standardized cesium carrier solution was added to an aliquot of the plutonium sample. All metallic elements were converted to perchlorate salts by addition of $HClO_4$ and heating to dense $HClO_4$ fumes. Cesium perchlorate ($CsClO_4$) was precipitated from this solution by depressing its solubility with the addition of absolute ethyl alcohol at ice bath temperatures. The solution was centrifuged and the supernatant decanted and discarded. The precipitate was washed with absolute ethyl alcohol several times and then filtered onto a preweighed glass-fiber disk, vacuum dried, and weighed on a Cahn electrobalance to determine the chemical yield. Counting of the $^{137}CsClO_4$ was again done by Ge(Li) counting of the 0.662 MeV gamma ray, but with a system of lower detector resolution and fewer channels over the region of interest. For this reason an absolute standard of $^{137}CsClO_4$ was prepared in an identical manner to the $^{137}CsClO_4$ separated from the plutonium samples, and the 0.662 MeV gamma activity of the standard compared directly to the unknown samples. The absolute ^{137}Cs standard was obtained from Nuclear Chicago Corporation and was prepared under the auspices of the ASTM Committee on Burnup and standardized by the mass spectrometric isotope dilution technique. Fissions were calculated by converting the ^{137}Cs (0.662 MeV) activity to atoms by means of the standard and applying the appropriate ^{239}Pu fission yield.

Target atoms. Since the plutonium samples were originally loaded in the capsules as a wet nitrate, the total number of target atoms present was determined after the irradiation by means of solid-state alpha spectrometry. This technique required good chemical separation from the stainless steel capsule components, primarily iron. The previously described solvent-extraction procedure provided this separation. However, to determine the chemical yield of the plutonium in the solvent-extraction separation, an internal ^{238}Pu tracer was introduced to an aliquot sample before any separations were made. Mass spectrometric analysis gave the following composition for the ^{238}Pu tracer: 81.2 at. % ^{238}Pu, 15.6 at. % ^{239}Pu, 2.6 at. % ^{240}Pu, 0.5 at. % ^{241}Pu, and 0.1 at. % ^{242}Pu. This composition results in an activity ratio of 1.4×10^3 for $^{238}Pu/^{239}Pu$; the alpha

activity contributed by ^{240}Pu and ^{241}Pu was negligible. The disintegration rate per unit volume of the ^{238}Pu tracer was calibrated by three independent means. First, an aliquot from a known volume was plated on a platinum disk, and the 5.50 MeV peak counted alpha spectrometrically using a calibrated silicon detector. The detector was calibrated by counting an absolute ^{241}Am standard obtained from Amersham Radiochemical Center. Secondly, another aliquot was pipetted into a PPO-POPOP-dioxane Napthalene liquid scintillation solution and counted on a Packard Tricarb liquid scintillation counter, whose counting efficiency is 100%. Thirdly, an aliquot was mounted on platinum and counted in a 2π-α proportional counter which had been previously calibrated by low-geometry alpha counting. The three methods resulted in measured disintegration rates per milliliter for ^{238}Pu which differed by less than 1%. The amount of ^{238}Pu activity added to the samples was estimated to be the same magnitude as that of the ^{239}Pu in the samples. To insure isotopic exchange between the samples and the tracer, the samples were fumed with $HClO_4$ to incipient dryness. The visual observation that the chromium in the sample had been converted to dichromate indicated that all the plutonium had been oxidized to the hexavalent state. The samples were allowed to cool and the residues taken up in HNO_3 and $Al(NO_3)_3$. The plutonium was extracted into methyl isobutyl ketone, and aliquots of the organic phase mounted on platinum for counting. The chemical yield (typically 96%) was determined from the observed activity under the 5.50 MeV peak of ^{238}Pu; the number of atoms of ^{239}Pu was determined from the activity under the 5.15 MeV peak of ^{239}Pu, after correction for chemical yield.

3. Uranium-235

Dissolution. In view of the dissolution difficulties encountered with the ^{233}U and ^{239}Pu samples, a slightly different approach was taken for the ^{235}U samples. Because the procedure involved the use of HF, the samples were dissolved in Teflon beakers equipped with covers. The samples were treated with a 4:1 HCl-HNO_3 mixture until only the silicate residue and a few dark metallic specks remained; HF was then added and the solution gently heated to volatilize silicon as fluosilicic acid. Upon cooling, it was apparent the metallic residue was still undissolved; therefore, more aqua regia and $HClO_4$ were added and the solution heated to fumes of $HClO_4$. The solution was cooled, 8N HNO_3 and more HF were added, and it was heated again until $HClO_4$ fuming. After this treatment the dissolution appeared to be complete, and the clear solution was transferred to a volumetric flask and made up to volume with 8N HNO_3.

Captures. The ^{236}U capture product was determined by mass spectrometric analysis. Separation from the stainless steel components was by solvent extraction (described in the analysis of the capture product of ^{239}Pu).

In addition to the unknown samples and the encapsulated control sample, a sample of the as-received $^{235}U_3O_8$ was also submitted for mass analysis to ascertain whether or not any contamination from natural uranium had occurred during dissolution or separation. The good agreement between results from the two control samples confirmed that no contamination had occurred during the chemical manipulations.

Fissions. Fissions were determined by $CsClO_4$ precipitation of the ^{137}Cs fission product, as in case of the ^{239}Pu samples.

Target Atoms. Initial ^{235}U target atoms were determined from the accurately weighed amounts of U_3O_8 prior to irradiation. Final uranium atoms were calculated from the initial atoms minus the number of atoms fissioned and captured, as in the case of the ^{233}U samples.

4. Plutonium-240

Dissolution. Dissolution of the ^{240}Pu samples was done identically to that described for ^{235}U using aqua regia, HF, and $HClO_4$. Unlike the ^{239}Pu samples, no undissolvable residue was detectable.

Captures. The ^{241}Pu capture product was determined by mass spectrometric analysis. Samples were prepared for this analysis similarly to the ^{239}Pu and ^{235}U samples.

Fissions. Fissions were determined by $CsClO_4$ precipitation of the ^{137}Cs as described in the ^{239}Pu separation.

Target Atoms. Final target atoms were determined by solid-state alpha spectrometry using the ^{238}Pu tracer technique described in the analysis of the ^{239}Pu samples.

5. Uranium-238

Dissolution. These samples were dissolved similarly to the ^{235}U samples.

Captures. The long lived ^{239}Pu capture product was determined by 2π-α proportional counting. An aliquot of the solution was pipetted directly onto a 2-in. dia. counting disk and allowed to spread over the entire area to reduce self-absorption of the 5.15 MeV ^{239}Pu alpha particles. From the estimated burnup of the samples, the amount of ^{239}Pu produced would result in a ^{239}Pu-to ^{238}U specific-activity ratio of 10^3-10^4. The 2π-α counts could therefore be considered to be due to ^{239}Pu alone.

Fissions. Fissions were determined identically to the samples of ^{239}Pu and ^{235}U.

Target Atoms. Initial atoms and final atoms were determined similarly to the samples of ^{233}U and ^{235}U.

6. Plutonium-242

Dissolution. These samples were dissolved similarly to the samples of ^{235}U, ^{240}Pu, and ^{238}U.

Captures. The ^{243}Pu capture product (4.98 h) decays to ^{243}Am (7.9×10^3 y) which in turn decays to ^{239}Np (2.36 d). Because a secular equilibrium condition exists between ^{243}Am and ^{239}Np, the number of captures of ^{242}Pu was determined by Ge(Li) counting of the gamma rays of ^{239}Np (209, 228, and 278 keV). A correlation between ^{243}Am atoms and gamma counts

under these three peaks was made by the utilization of a ^{243}Am-^{239}Np standard. Calibration of this standard consisted of first separating ^{239}Np from ^{243}Am by a liquid ion-exchange method.[11] Tricaprylyl methyl ammonium nitrate dissolved in xylene was used for the quantitative extraction of the ^{239}Np. The acid concentration of the standard was 5N HNO_3; this solution was made 0.01N in $NaNO_2$ to stabilize the neptunium in the +4 state. After the addition of $NaNO_2$, the organic phase was added (organic/aqueous = 0.5) and the mixture shaken vigorously for about 30 sec. The phases were allowed to separate for about an hour before the organic phase containing the neptunium was removed. Fresh organic solution was added and the process repeated. Finally, the aqueous solution was scrubbed twice with half volumes of xylene to remove any residual quaternary salt. The organic phases were combined for Ge(Li) counting of the 209, 228, and 278 keV gamma rays of ^{239}Np. The aqueous phase was made up to a known volume and aliquots taken for alpha spectrometry and absolute calibration. The solid-state alpha-spectrometric analysis indicated 86.4% ^{243}Am, 10.5% ^{244}Cm, 2.1% ^{241}Am. The absolute calibration was done by liquid scintillation counting and as a check, 2π-α counting. The total alpha disintegration rate was corrected by means of the alpha-spectrometric ratio to obtain the disintegration rate due to ^{243}Am alone. This disintegration rate was converted to atoms of ^{243}Am and correlated with the ^{239}Np gamma counts.

The irradiated ^{242}Pu samples required the elimination of all traces of perchlorate and the conversion of all salts to the nitrate in preparation for extraction. When this was accomplished, the samples were treated by the separation procedure described above, and the organic phase, containing both neptunium and plutonium, was discarded. The aqueous phase containing only ^{243}Am was allowed to come to equilibrium with the ^{239}Np for a period of three weeks. At the end of this period, the ^{239}Np was again separated from the ^{243}Am by the extraction procedure, and the ^{239}Np gamma counted as described. These gamma counts were then correlated to the ^{243}Am-^{239}Np standard and the number of captures calculated.

Fissions. The total number of cesium atoms was determined by $CsClO_4$ precipitation of ^{137}Cs, as previously described. However, the calculation of the number of fissions due to ^{242}Pu alone is difficult because of the isotopic impurity of the target material. Fissioning of ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Pu also contribute to the observed number of cesium atoms and their contribution varies with position in the reactor. A description of how these corrections were made is given in the section on error analysis.

Target Atoms. Target atoms of ^{242}Pu were determined by solid-state alpha spectrometry using the ^{238}Pu impurity as an internal standard. Since ^{238}Pu was the major alpha contributor in the samples and mass ratios of all plutonium constituents were known from the control sample, the ^{238}Pu disintegration rate could be correlated with the number of atoms of ^{242}Pu. Mass spectrometric results before and after irradiation indicated that the 238/242 ratio changed by about 1%, which was insignificant to the calculation. A chemical separation of the plutonium was required before counting because of the significant amount of ^{241}Am present due to the ^{241}Pu impurity. For this reason an internal tracer of high purity ^{239}Pu was added with an activity level 5-10 times greater than the ^{238}Pu present in order to reduce errors in integration of the respective alpha peaks.

The isotopic composition of the ^{239}Pu was 99.97% ^{239}Pu and 0.003% ^{240}Pu; this solution was calibrated by liquid scintillation counting and checked by 2π-α counting. A known amount of the ^{239}Pu tracer was added to the ^{242}Pu samples and both were fumed with $HClO_4$ to insure exchange and oxidation to the hexavalent state. Several ferric hydroxide scavenges were done on the cooled solution to insure complete removal of any dichromate interference due to the stainless steel. When a colorless supernatant remained, indicating dichromate had been successfully removed, the plutonium-bearing ferric hydroxide was dissolved in nitric acid and the solution diluted with a saturated solution of ammonium nitrate. The plutonium was extracted into methyl isobutyl ketone, the phases allowed to separate overnight, and the organic phase plated on platinum for alpha spectrometry. The 5.15 MeV peak of ^{239}Pu was used to determine the chemical separation and the 5.50 MeV peak of ^{238}Pu was correlated with the mass spectrometric results of the control sample to determine the ^{242}Pu atoms.

III. Error Analysis

The uncertainties associated with the results of this study can, in general, be categorized into two groups: uncertainties related to chemical analysis and uncertainties related to counting. Under the former, consideration was given to errors in pipetting of aliquots, chemical separation, weighing, and mass spectrometric determination. Any one of these did not exceed ±1%. Counting considerations included errors in calibration of detectors, integration of counts under the particular peak of interest, calibration of tracers, and correlations between standards and unknowns. The largest single error in this group was the uncertainty in the number of cesium atoms in the ^{137}Cs Nuclear Chicago standard, which was ±2.1%. All other errors were ±1% or less. The errors assigned to determining captures were ±1.1% for ^{233}U and ±1.9% for ^{238}U. Errors in the fission rates were ±2.9% for ^{233}U and ±3.0% for both ^{235}U and ^{238}U. The error in alpha was ±3.1% for ^{233}U and ^{235}U, ±3.4% for ^{238}U, and ±3.1% for ^{239}Pu.

In the case of ^{240}Pu fissions, an additional correction was required due to the contribution of ^{239}Pu fission to the observed ^{137}Cs atoms. This was arrived at by using the experimental ^{137}Cs atoms/^{239}Pu atom ratio obtained from the ^{239}Pu samples in the same reactor position and correcting for the amount of ^{240}Pu determined by the mass spectrometric analysis. The uncertainty in this correction resulted in ±0.5% uncertainty in ^{240}Pu fissions. The total error in ^{240}Pu fissions was 3.5%. The error in captures and alpha was 2.1% and 3.1% respectively.

The number of fissions occurring in the ^{242}Pu samples required several corrections because of the contribution due to ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Pu fissions. The assumptions made in order to calculate these corrections were as follows: (1) Initially, the ^{242}Pu capture-to-fission ratio was considered to be equal to the experimentally determined ^{240}Pu capture-to-fission ratio. After all isotopic corrections were made, this assumption was iteratively adjusted to be internally consistent in the calculation. (2) The ratio of ^{241}Pu $\sigma_f/^{239}Pu$ $\sigma_f = 1.2 \pm 0.2$ at the core position and increases to 1.9 ± 0.3 in the blanket regions. (3) The ratio of ^{239}Pu $\sigma_f/^{238}Pu$ $\sigma_f = 1.5 \pm 0.5$ at all positions within the reactor. Utilizing these assumptions, the number of fissions for the impurities was calculated, appropriate ^{137}Cs yields applied, and this contribution subtracted

from the total number of fissions determined by counting $^{137}CsClO_4$. In positions that were greater than 30 cm from core center, all the observed fissions were considered to be from impurities and data for ^{242}Pu beyond 30 cm were not analyzed. The errors assigned to the results listed represent primarily the contribution due to the errors assigned to the assumptions made; however, all error considerations were made. The total error on captures was ±3.1% for both the core and blanket positions; whereas, the total error on fissions ranged from ±4.4% in the core to ±13.2% in the blanket; and similarly, the error on alpha ranged from ±4.1% in the core to ±13.3% in the blanket.

IV. RESULTS

Presented in Tables 5-10 are capture rates, fission rates, and alpha values as a function of reactor position. Row number refers to the position of the subassembly in the reactor, and the radial and axial distances given are in centimeters measured from the vertical center line and horizontal mid-plane, respectively, of the reactor to the actual location of the sample. Negative axial distances refer to samples which were below the horizontal mid-plane. Exposure listed is the total accumulated sample exposure expressed in megawatt-days. Capture and fission rates are expressed in units of atoms produced per target atom per megawatt day. The variation of alpha (captures/fissions) for the fissile and fertile isotopes as a function of reactor position is illustrated in Figures 3 through 6. Errors, in general, are represented by the size of each datum point, with the exception of ^{242}Pu, and were assigned in the manner already discussed.

V. DISCUSSION

A thorough discussion of these results cannot be made until appropriate neutronic calculations at each of the 15 irradiation locations have been performed. As yet, these calculations have not been completed; at the appropriate time, this analysis will be presented.

The following general comments, concerning the results as they are presented here, should be recognized. Errors assigned to the capture rates in Tables 5-10 represent the absolute errors. Errors assigned to fission rates and the alpha values include all known uncertainties exclusive of uncertainties in the ^{137}Cs fission yield. No errors have been assigned to the fission rates and capture rates of the ^{239}Pu samples reported in Table 8. This is because of the difficulties encountered in determining the initial number of ^{239}Pu atoms present in each of the capsules. (The fission and capture rates are dependent on this quantity; however, the capture-to-fission ratios are not.) As described earlier, the ^{239}Pu starting material was not stoichiometric and as a result had to be loaded into the capsules as a nitrate solution. The solution was allowed to dry within the capsule and the capsule was then capped and welded, with the intent that the ^{239}Pu atoms contained would be radiochemically determined after the irradiation.

Dissolutions of the irradiated ^{239}Pu capsules were complicated by an insoluble black residue, which was impervious to various acid treatments. Gamma counting of this residue indicated that no ^{137}Cs was present and alpha counting indicated that no significant ^{239}Pu was present. Spark-

TABLE 5. Capture Rates, Fission Rates, and Alpha for ^{233}U

Reactor Position			Exposure	Capture Rate	Fission Rate	Alpha
Row	Radial (cm)	Axial (cm)	MWd	(10^{-7} atoms/ atom-MWd)	(10^{-6} atoms/ atom-MWd)	(Captures/Fissions)
1	2.50	51.5	3069	1.512±0.017	1.454±0.043	0.1040±0.0032
1	2.86	17.0	3069	5.471±0.061	6.964±0.205	0.0786±0.0024
1	2.86	-17.0	3069	5.763±0.065	7.325±0.216	0.0787±0.0024
1	2.86	8.5	3069	6.949±0.078	9.384±0.277	0.0741±0.0023
1	2.86	-8.5	3069	6.980±0.078	9.487±0.280	0.0736±0.0023
1	2.86	0.0	3069	7.488±0.084	10.293±0.304	0.0728±0.0022
5	21.25	-3.2	4293	5.531±0.062	7.763±0.229	0.0713±0.0022
9	40.83	-2.3	7640	1.937±0.022	2.040±0.060	0.0950±0.0029
13	61.24	-2.3	9985	0.3564±0.004	0.3366±0.010	0.1059±0.0033

TABLE 6. Capture Rates, Fission Rates, and Alpha for ^{235}U

Reactor Position			Exposure	Capture Rate	Fission Rate	Alpha
Row	Radial (cm)	Axial (cm)	MWd	(10^{-7} atoms/ atom-MWd)	(10^{-6} atoms/ atom-MWd)	(Captures/Fissions)
1	2.86	72.9	3069	1.506±0.017	0.3875±0.011	0.3887±0.0119
1	2.86	51.5	3069	4.098±0.046	1.113±0.033	0.3682±0.0113
1	2.86	30.2	3069	7.513±0.084	2.963±0.087	0.2536±0.0078
1	2.86	17.0	3069	9.495±0.106	4.662±0.138	0.2037±0.0063
1	2.86	-17.0	3069	10.199±0.114	4.798±0.142	0.2126±0.0065
1	2.86	-8.50	3069	11.586±0.130	6.337±0.187	0.1828±0.0056
1	2.86	0.0	3069	12.441±0.139	6.930±0.204	0.1795±0.0055
3	10.21	-3.50	3069	11.576±0.130	6.332±0.187	0.1828±0.0056
7	30.62	-2.30	2035	6.843±0.077	2.977±0.088	0.2299±0.0071
11	51.03	-2.30	7640	1.672±0.019	0.5754±0.017	0.2906±0.0089
15	71.45	-2.30	9985	0.3120±0.0035	0.0926±0.0027	0.3369±0.0103

TABLE 7. Capture Rates, Fission Rates, and Alpha for ^{238}U

Reactor Position			Exposure	Capture Rate	Fission Rate	Alpha
Row	Radial (cm)	Axial (cm)	MWd	(10^{-7} atoms/ atom-MWd)	(10^{-8} atoms/ atom-MWd)	(Captures/Fissions)
1	2.86	17.0	3069	4.946±0.092	26.644±0.796	1.857±0.063
1	2.86	-17.0	3069	5.646±0.106	25.540±0.753	2.211±0.075
1	2.86	8.5	3069	6.432±0.120	44.148±1.302	1.457±0.049
11	51.03	1.78	7640	0.8365±0.016	0.2474±0.0073	33.81±1.15
15	71.45	1.78	9985	0.1540±0.0029	0.0208±0.0006	73.89±2.51

TABLE 8. Capture Rates, Fission Rates, and Alpha for ^{239}Pu

Reactor Position			Exposure	Capture Rate	Fission Rate	Alpha
Row	Radial (cm)	Axial (cm)	MWd	(10^{-7} atoms/ atom-MWd)	(10^{-6} atoms/ atom-MWd)	(Captures/Fissions)
1	2.50	72.9	3069	1.932	0.4274	0.4520±0.0139
1	2.86	30.2	3069	7.179	3.368	0.2132±0.0065
1	2.86	17.0	3069	7.065	5.782	0.1222±0.0038
1	2.86	-17.0	3069	7.813	5.814	0.1344±0.0041
1	2.86	8.5	3069	7.796	7.568	0.1030±0.0032
1	2.86	-8.5	3069	7.955	8.576	0.0928±0.0028
1	2.86	0.0	3069	8.481	9.977	0.0850±0.0026
3	10.21	0.32	3069	7.915	9.672	0.0818±0.0025
5	21.25	0.32	4293	8.969	9.111	0.0984±0.0030
7	30.62	0.42	2035	4.856	3.624	0.1340±0.0041
9	40.83	0.42	7640	2.720	1.901	0.1431±0.0044
11	51.03	0.42	7640	1.260	0.6828	0.1845±0.0057
13	61.24	0.42	9985	0.565	0.2344	0.2414±0.0074
15	71.45	0.42	9985	0.315	0.0856	0.3680±0.0113

TABLE 9. Capture Rates, Fission Rates, and Alpha for ^{240}Pu

Reactor Position			Exposure	Capture Rate	Fission Rate	Alpha
Row	Radial (cm)	Axial (cm)	MWd	(10^{-7} atoms/ atom-MWd	(10^{-6} atoms/ atom-MWd)	(Captures/Fissions)
1	2.50	72.9	3069	4.230±0.087	0.0205±0.0006	20.634±0.633
1	2.50	51.5	3069	3.324±0.069	0.1106±0.0033	3.005±0.092
1	2.50	30.2	3069	7.795±0.161	0.6831±0.0202	1.141±0.035
1	2.86	8.5	3069	10.945±0.225	3.162±0.093	0.3461±0.0106
1	2.86	0.0	3069	10.415±0.215	3.419±0.101	0.3046±0.0094
3	10.21	2.22	3069	9.575±0.197	3.111±0.092	0.3078±0.0095
5	21.25	2.22	4293	7.807±0.161	2.220±0.065	0.3517±0.0108
7	30.62	1.78	2035	5.894±0.121	0.9916±0.0293	0.5944±0.0183
9	40.83	1.78	7640	3.177±0.065	0.2320±0.0068	1.369±0.042
13	61.24	1.78	9985	0.7464±0.0154	0.0218±0.0006	3.424±0.105

TABLE 10. Capture Rates, Fission Rates, and Alpha for ^{242}Pu

Reactor Position			Exposure	Capture Rate	Fission Rate	Alpha
Row	Radial (cm)	Axial (cm)	MWd	(10^{-7} atoms/ atom-MWd)	(10^{-7} atoms atom-MWd)	(Captures/Fissions)
1	2.50	79.9	3069	1.465±0.045	a	-
1	2.50	51.5	3069	1.755±0.054	a	-
1	2.86	30.2	3069	4.418±0.137	1.126±0.149	3.924±0.522
1	2.86	17.0	3069	4.812±0.149	5.950±0.523	0.8087±0.0728
1	2.86	8.5	3069	4.993±0.155	10.488±0.916	0.4761±0.0425
1	2.86	-8.5	3069	5.399±0.167	10.4880±0.950	0.4962±0.0443
1	2.86	0.0	3069	5.446±0.169	11.614±0.512	0.4689±0.0193
3	10.21	-1.59	3069	5.777±0.179	12.064±1.053	0.4789±0.0428
5	21.25	-1.59	4293	4.216±0.131	7.362±0.644	0.5727±0.0513
7	30.62	-0.94	2035	3.037±0.094	2.111±0.279	1.439±0.191
9	40.83	-0.94	7640	1.935±0.060	a	-
11	51.03	-0.94	7640	0.783±0.024	a	-
13	61.24	-0.94	9985	0.387±0.012	a	-
15	71.45	-0.94	9985	0.243±0.008	a	-

[a]Not calculated because of isotopic impurities (see text).

Figure 3. Fissile Isotopes: Alpha vs. Radial Position

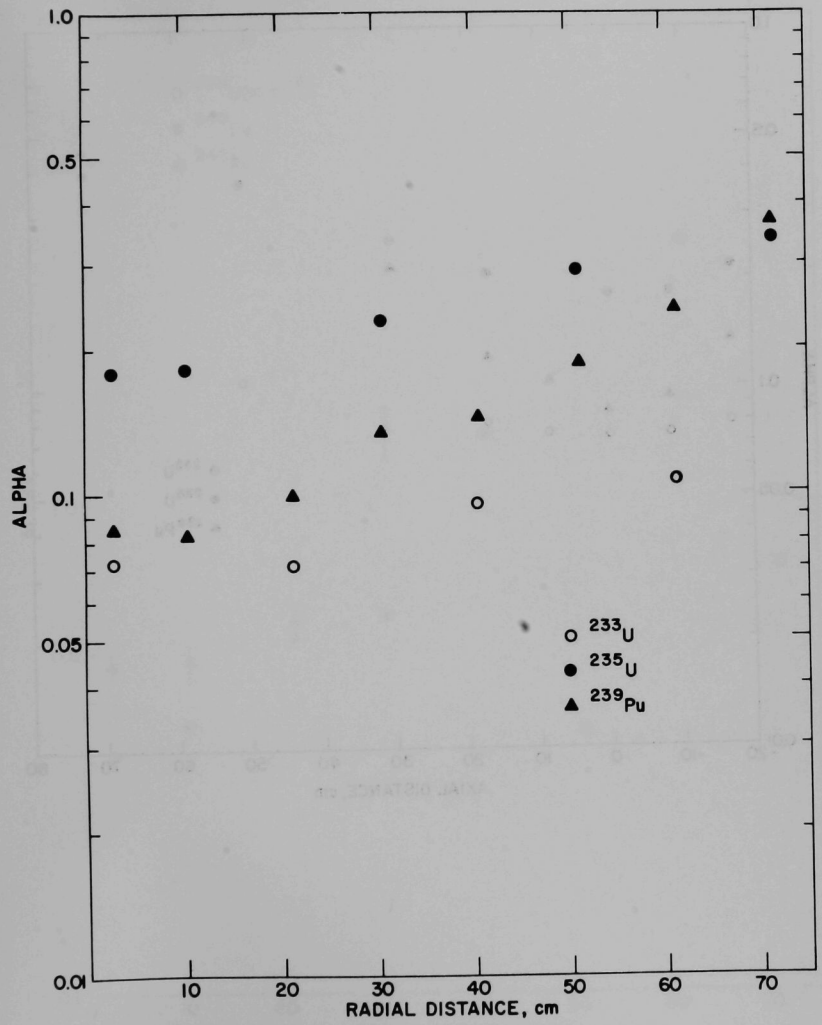


Figure 4. Fissile Isotopes: Alpha vs. Axial Position

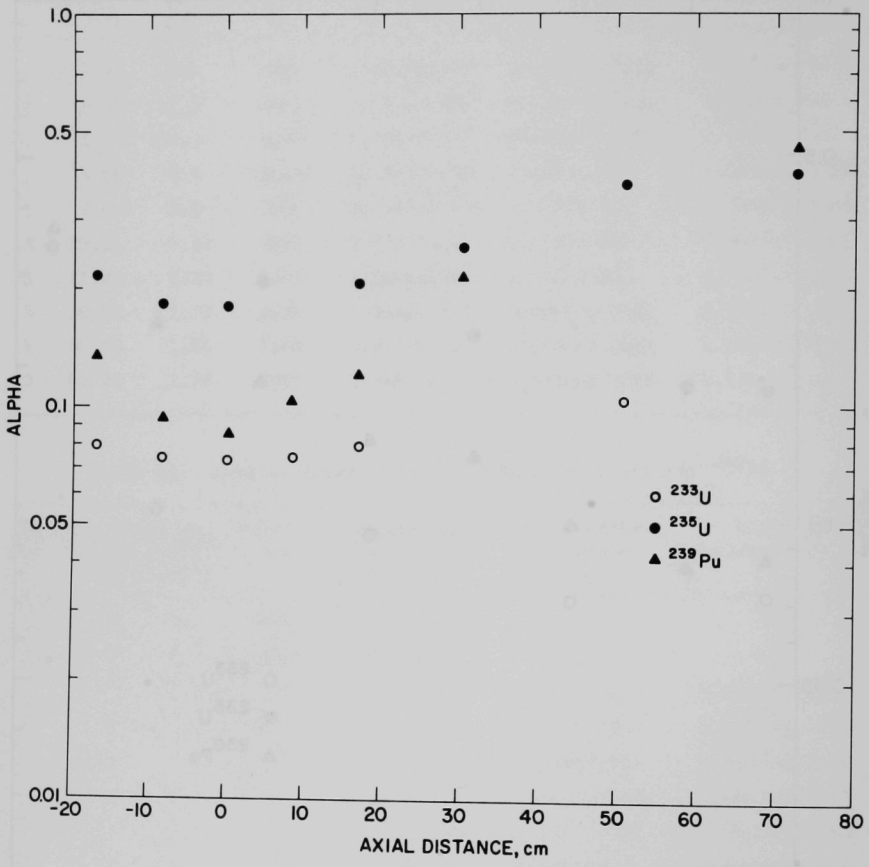


Figure 5. Fertile Isotopes: Alpha vs. Radial Position

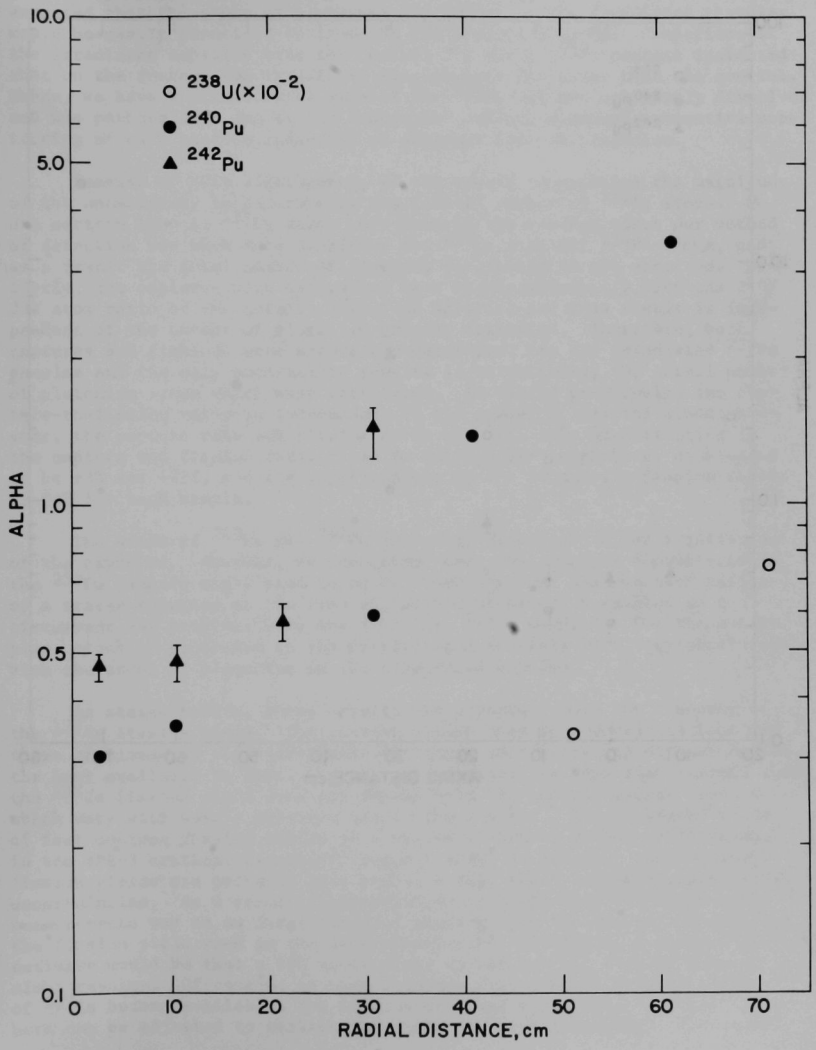
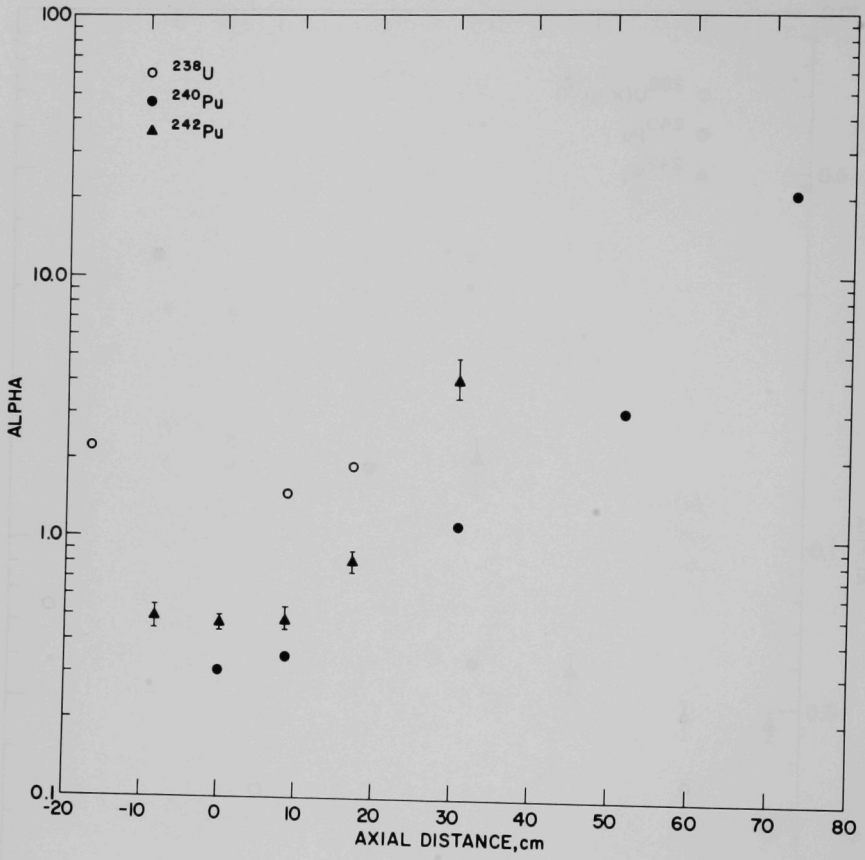


Figure 6. Fertile Isotopes: Alpha vs. Axial Position



source mass spectrometry showed the residue to be primarily carbon. The unirradiated ^{239}Pu control sample dissolved normally and no trace of residue was detected. Each capsule, including the unirradiated control, was originally loaded with the same volume of $Pu(NO_3)_x$ solution and it was expected that the atoms of plutonium determined in the irradiated capsules would be nearly identical to those in the control capsule. Comparison of the irradiated capsules with the control for total ^{239}Pu content indicated that on the average the irradiated samples were 23% lower than the control. Hence, we have concluded that some of the ^{239}Pu was not completely dissolved and was perhaps lost due to the insoluble residue, although exhaustive monitoring of each residue indicated no apparent loss of plutonium.

Because of this discrepancy, we are unable to appraise the magnitude of the uncertainty in determining the initial number of ^{239}Pu atoms. We are certain that no ^{137}Cs atoms were lost in the residue since our method of detection was much more sensitive for ^{137}Cs than for ^{239}Pu alone, and as a result the total number of fissions determined is not affected. Similarly, the captures were determined mass spectrometrically from the 240/239 atom ratio of the totally dissolved material and this result is independent of the amount of plutonium present initially. Therefore, both captures and fissions were accurately determined for the irradiated ^{239}Pu samples and the only uncertainty results in establishing the actual number of plutonium atoms which were irradiated. As stated previously, the capture-to-fission ratio is independent of the number of initial atoms; however, the capture rate and fission rates are not. The uncertainties in the capture and fission rates given in Table 8 are, therefore, considered to be +3% and -23%, and the uncertainties in the capture-to-fission ratios ±3.04% for each sample.

The atoms of ^{240}Pu and ^{242}Pu were also determined after irradiation of the capsules. However, we recognized that the problems encountered in the ^{239}Pu samples might also exist in these samples, and the utilization of a tracer solution at the time of capsule dissolution enabled us to circumvent any problems with the ^{240}Pu and ^{242}Pu samples. For these samples, atoms of plutonium in the unirradiated controls were very consistent with the atoms of plutonium in the irradiated samples.

As stated before, these results are dependent upon the accuracy of the ^{137}Cs fission yield. The fission yields used in our calculations are those of Lisman et al[10] (see Table 4); these values are, in our estimation, the best available to date. In the calculations, we have also assumed that the ^{137}Cs fission yield does not depend upon the neutron-energy spectra which vary with sample position within the reactor. Recent measurements of fast-neutron fission yields in a series of EBR-II mockup irradiations in the ZPR-3 critical assembly[12] suggest that, in general, fast-neutron fission yields are probably less reliable than their reported experimental uncertainties. As a result, uncertainties in fast-reactor fission-rate measurements may be as large as 12% depending upon the fission product and the fission yield used in the determination.[12] Therefore, a conservative estimate would be that a 10% uncertainty exists for all fission rate and alpha results. Of course, as more accurate fast-neutron fission yields of ^{137}Cs become available, the fission-rate and alpha results reported here can be adjusted to reflect this more accurate parameter. The uncer-

tainties that we have assigned to the results in Tables 5-10 are realistic assessments of the relative values for each of the heavy elements. Thus, changes in fission rates and alpha as a function of reactor location have indeed been determined to the quoted accuracies.

One of the more important quantities that can be determined from the measurement of alpha is the breeding potential of the heavy elements. The breeding potential, B, is defined as

$$B = \frac{(\nu - 1 - \alpha)}{1 + \alpha}$$

where ν is the number of neutrons emitted per fission of the heavy element and α is the capture-to-fission ratio. From the measured alpha values and ν values from reference 13, we have computed the average breeding potential for the six heavy isotopes studied. These results are summarized in Table 11. Crouthamel et al[14] have previously reported similar results for ^{233}U, ^{235}U, and ^{239}Pu from EBR-I irradiations. The breeding potentials for these isotopes in the core of EBR-I are identical to the values determined from this work for EBR-II core center. The EBR-II results indicate that the breeding potential for ^{233}U (1.4) varies by less than 5% throughout the reactor, whereas for ^{239}Pu the breeding potential is 1.7 in the core and drops to 1.1 at a radial or axial distance of 70 cm. For ^{235}U the breeding potential is slightly greater than unity in the core, and decreases to slightly less than unity outside the core. Of the three fertile isotopes, ^{240}Pu provides the largest number of neutrons for breeding within the core where the neutron energy is highest; however, its breeding potential is very sensitive to neutron energy and drops to -0.6 in the blanket region. The breeding potential of ^{238}U is zero or negative for all spectra measured.

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TABLE 11. Average Breeding Potential of Uranium and Plutonium Isotopes in EBR-II

Nuclide	Reactor Position	Average Breeding Potential
^{239}Pu	Core {<18 cm axial, <24 cm radial}	1.7
	Blanket (>24 cm)	1.6 to 1.1
^{233}U	Core	1.4
	Blanket	1.3
^{235}U	Core	1.1
	Blanket	1.0 to 0.9
^{240}Pu	Core	1.4
	Blanket	0.7 to -0.6
^{242}Pu	Core	1.1
	Blanket	0.0
^{238}U	Core	0.0
	Blanket	-0.9

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